



Effect of photo-induced charge separated state lifetimes in donor-acceptor1-acceptor2 organic ambipolar semiconductors on their photovoltaic performances



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ABSTRACT

A newly ambipolar organic semiconductor with styrene based indoline derivative (YD) as the electron donor (D), s-triazine group (TRC) as the electron acceptor (A₁), and fullerene derivative (NMF) as the second electron acceptor (A₂) has been synthesized and characterized, which had a charge-separated state of 7.9 μs, corresponding to a 37 fold increase in comparison with the 215 ns lifetime of the D-A₁ precursor. Here we compared photovoltaic properties of YD-TRC-NMF with those of other four D-TRC-A₂ systems, which replaced indoline with triphenylamine (MTPA) and fullerene with perylene bisimide or anthraquinone (AEAQ), indicating that long-lived CS states are playing an important role in promoting photoelectric conversion in YD-TRC-AEAQ (or NMF) and MTPA-TRC-AEAQ, in which the short-circuit current and photoelectric conversion efficiency were significantly improved in comparison with YD-TRC and MTPA-TRC, respectively, due to triplet states of A₂ being higher than charge-separated states. This proves that a general D-A₁-A₂ architecture can become a reasonable materials design strategy for photovoltaics.

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1. Introduction

Natural photosynthesis, the most efficient process of converting sunlight into electrical or chemical energy, is of great interest to researchers focused on energy conversion and utilization. In a natural photosynthetic system, long-lived charge-separated (CS) states are formed via multistep electron transfers upon light

absorption and are believed to be the key for the subsequent chemical reactions [1–3]. Great efforts have been made to design dyes in which long-lived CS states are attained through photo-induced electron transfer [4–9]. In several of the artificial photosynthetic complexes, photo-induced CS states have reached the lifetimes of 380 ms to 2 h that are comparable to, or even longer than those in nature photosynthetic systems [5,7]. It is recognized that materials with long-lived CS states may improve the performance of organic solar cells (OSCs) [10–13]. Actually, generation of CS states with suitable lifetimes is critical in the formation of photocurrents for solar cells [14–17], that means the study of photoinduced charge separation processes and investigation of their essential role in improving the efficiency of solar cell devices should be required in depth. However, only a few publications have focused on the application of long-lived CS organic compounds to OSCs. It is important to perform systematic studies to establish a relationship between the lifetimes of CS states and the photovoltaic

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performance of OSCs. This contains two levels of work: 1) Design and construction of organic materials which had long-lived CS states; 2) Select the appropriate structure of OSCs device to investigate the effect of long-lived CS states of the material effectively.

Photo-induced CS were discovered in organic systems containing donor (D) and acceptor (A) moieties via photo-induced intramolecular electron transfer. This result suggests that D and A determine the driving force of electron transfer [18–20] while the distance, spatial orientation, and flexibility between D and A have a marked impact on the rate of photo-induced electron transfer and the efficiency of generating CS species and lifetimes of CS states. Photo-induced CS states can also be created by photo-induced hole transfer when the acceptor moiety is initially photo-excited [19]. That means that ambipolar organic semiconductors, which include both photo-induced electron and hole transfers, can form CS states effectively because of the synergistic combination of above two transfers [21–28]. Using a D-A₁-A₂ strategy, we reported an ambipolar organic semiconductor MTPA-TRC-AEAQ using *s*-triazine (TRC) as the first acceptor to connect the donor styrene based triphenylamine (MTPA) with anthraquinone (AEAQ) as the second acceptor, which had been proven to be a good linker to promote the photo-induced electron transfer and increase the light-stability for MTPA-TRC-AEAQ [29,30]. Upon the absorption of photons, the CS states in the organic semiconductor MTPA-TRC-AEAQ was elongated to 650 ns, more than eight-fold compared to those in the D-A₁ precursor MTPA-TRC (80 ns). MTPA-TRC exhibits similarly low photovoltaic characteristics in the single layer OSCs with common organic semiconductors such as phthalocyanine, poly(phenylenevinylene) and triarylamine derivatives [12,31–33], of which the efficiency has been improved to greater than 6% and can even exceed 10% by the combined application of other semiconductors and the proper design of solar cell structures [12,34–39]. Importantly, the single layer OSCs constructed using MTPA-TRC-AEAQ were found to be significantly more efficient than those using MTPA-TRC [29], indicating that single layer OSCs had been proven to be the highly effective cell structure reflecting the effect of long-lived CS states of organic materials directly. That was why we chose single-layer OSC structure in this paper, although its photoelectric conversion efficiency was relatively low in various literature [12,31–33,38,40–42].

Encouraged by these results, we obtained a new ambipolar organic semiconductor YD-TRC-AEAQ by substitution of MTPA for an indoline derivative (YD) which has been reported to be a more effective donor [29,43,44]. The lifetime of CS states reached 1.14 μs for YD-TRC-AEAQ [44]. Further studies suggest that charge separation efficiency and lifetime of CS states may be controlled by changing the donor and second acceptor of MTPA-TRC-AEAQ [29,43–45]. The aforementioned ambipolar organic semiconductors have cascade energy levels and similar photophysical processes to generate photo-induced CS states. Therefore, it is possible and significant to relate their CS states to the photovoltaic performance of single layer OSC devices, and reveals the advantage of introducing the acceptor2 to D-A₁ system to form the D-A₁-A₂ architecture in the research perspective of photovoltaic characteristics.

To systematically study the solar cell performance of these promising materials and to establish correlation between the lifetime of charge separation and cell efficiency, more systems are required. Therefore, we have synthesized a new ambipolar organic semiconductor, YD-TRC-NMF, introducing a fullerene derivative (NMF) as the second acceptor (Figs. S1–S2 of Electronic Supplementary Information (ESI)) to the reported D-A₁ system YD-TRC [43,44]. We investigated its photophysical properties by absorption and emission spectra, which showed that YD-TRC-NMF can

generate the CS state with lifetime of 7.9 μs. Then, we fabricated single layer OSCs devices with all of the above D-A₁-A₂ systems with their D-A₁ precursors. The photovoltaic properties were discussed in detail and were related to their charge separation processes and lifetime of CS state.

2. Experimental

2.1. Synthesis

Structures of the key compounds being synthesized and studied in this work are provided in Chart 1. While the other compounds were synthesized and studied previously [29,43–45], YD-TRC-NMF was first reported here and its synthetic routes were shown in Schemes 1 and 2. Other compounds discussed in this paper e.g. 4,4'-dimethyl-4''-styryl-triphenylamine (MTPA) [29] and 4-(4-methylphenyl)-7-styryl-1,2,3,3a,4,8b-hexahydrocyclopenta[b]indole (YD) [44](ESI, Chart S1), D-A systems 4,4'-dimethyl-4''-(4-(4,6-dichloro-1,3,5-triazin-2-ylamino)styryl)triphenylamine (MTPA-TRC) [29] and 4-(4-methylphenyl)-7-(4-(4,6-dichloro-1,3,5-triazin-2-ylamino)styryl)-1,2,3,3a,4,8b-hexahydrocyclopenta[b]indole (YD-TRC) [43,44], D-A₁-A₂ systems 4,4'-dimethyl-4''-(4-(4-chloro-6-(2-(9,10-dioxoanthracen-1-ylamino)ethylamino)-1,3,5-triazin-2-ylamino)styryl)triphenylamine (MTPA-TRC-AEAQ) [29], 4,4'-dimethyl-4''-(4-(4-Chloro-6-(N-(1-hexylheptyl)-N'-(4-amino)phenyl-perylene-3,4,9,10-tetracarboxylbisimide)-1,3,5-triazin-2-ylamino)styryl)triphenylamine (MTPA-TRC-PDI) [45], 4,4'-dimethyl-4''-(4-(4-Chloro-6-(N-(1-hexylheptyl)-N'-(4-amino)phenyl-1,7-di(4-*tert*-butylphenoxy)-perylene-3,4,9,10-tetracarboxylbisimide)-1,3,5-triazin-2-ylamino)styryl)triphenylamine (MTPA-TRC-PBI) [45] and 4-(4-methylphenyl)-7-(4-(4-chloro-6-(2-(9,10-dioxoanthracen-1-ylamino)ethylamino)-1,3,5-triazin-2-ylamino)styryl)-1,2,3,3a,4,8b-hexahydrocyclopenta[b]indole (YD-TRC-AEAQ) [44] were prepared according to the literature. The synthetic pathways of 4-(4-methylphenyl)-7-(4-(4-chloro-6-phenylamino-1,3,5-triazin-2-ylamino)styryl)-1,2,3,3a,4,8b-hexahydrocyclopenta[b]indole (YD-TRC-BA), *N*-methyl-2-(4-nitrophenyl)fulleropyrrolidine (NMF_n), *N*-methyl-2-(4-aminophenyl)fulleropyrrolidine (NMF_a), *N*-methyl-2-(4-(4,6-dichloro-1,3,5-triazin-2-ylamino)phenyl)fulleropyrrolidine (NMF_t), 4-(4-methylphenyl)-7-(4-(4-chloro-6-(*N*-methyl-2-(4-phenyl)-fulleropyrrolidine)ylamino-1,3,5-triazin-2-ylamino)styryl)-1,2,3,3a,4,8b-hexahydrocyclopenta[b]indole (YD-TRC-NMF) were illustrated in Schemes 1 and 2. All reagents and solvents were reagent grade and further purified by the standard methods if necessary. All synthetic procedures were carried out under an atmosphere of dry nitrogen unless otherwise indicated.

2.2. Synthesis of YD-TRC-BA

YD-TRC (77.2 mg, 0.15 mmol) was dissolved in anhydrous THF (20 mL) and then phenylamine (14.0 mg, 0.15 mmol) was added into the solution. The mixture was stirred at 40 °C for 6 h. Afterwards, the solvent was removed by rotary evaporation and the residue was purified by column chromatography on silica gel using dichloromethane as the eluent. An orange solid was yielded (21.6 mg, 25.2%). HRMS-ESI (*m/z*): 571.2373 [M+H]⁺ (calcd for C₃₅H₃₂ClN₆⁺: *m/z* = 571.2371). ¹HNMR (500 MHz, CDCl₃): δ 7.55–7.47 (m, 7H), 7.34 (s, 1H), 7.18 (m, 7H), 7.03 (d, *J* = 16 Hz, 1H), 6.89 (dd, *J* = 16, 8.5 Hz, 2H), 4.81 (m, 1H), 3.85 (m, 1H), 2.35 (s, 3H), 2.12–1.49 (m, 6H). Elem. Anal.: Found: C, 73.52; H, 5.40; N, 14.84% (Calc. for C₃₅H₃₁ClN₆: C, 73.61; H, 5.47; N, 14.72%)

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